



Evaluation of the carbon dioxide (CO₂) emission factor from lime applied in temperate upland soil

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ABSTRACT

Agricultural liming materials are broadly utilized in the world to ameliorate soil acidity. Crushed limestone (CaCO₃) and dolomite (MgCO₃·CaCO₃) are most common liming materials. The CO₂ emission coefficient was proposed as 0.12 Mg C per Mg for CaCO₃ by the Intergovernmental Panel on Climate Change (IPCC), which indicates that 100% of C in CaCO₃ is eventually released to the atmosphere in the form of CO₂. However, this assumption appears unlikely based on current knowledge of the very low solubility of CaCO₃ and carbonate transport through soil. To develop the direct CO₂ emission factor from the C-containing liming materials applied in moderately acidic soil, CaCO₃ was applied in a typical temperate upland soil at different levels (0–2 Mg ha⁻¹ yr⁻¹), and the CO₂ emission rates were determined using the closed chamber method for two years. The direct CO₂ emission factor from CaCO₃ was estimated using ¹³C fluxes from ¹³C-CaCO₃ applied soils. Approximately 0.026 Mg C per Mg of CaCO₃ was emitted as CO₂ from CaCO₃ in cropping lands. This value was much lower than the IPCC default value (0.12 Mg C per Mg of CaCO₃). The current CO₂ emission coefficient was comparable with the United States Environmental Protection Agency (EPA) emission factor (0.046 Mg C per Mg of CaCO₃) from the agricultural field. If the EPA emission factor from the ocean (0.013 Mg C per Mg of CaCO₃) is added to the currently developed CO₂ emission factor of the inner agricultural field, the total CO₂ emission factor from lime may be close to 0.039 Mg C per Mg of CaCO₃ in the selected upland soil. Therefore, we propose that the current IPCC guideline value of the CO₂ emission factor from lime should be revised as 0.039 Mg C per Mg of CaCO₃ for Korean upland soils.

1. Introduction

Liming is a common agricultural practice to decrease soil acidity and increase crop productivity (Haynes and Naidu, 1998). Agricultural lime is usually applied as crushed limestone or dolomite in soil. In 2013, 46 Mg of limestone and 193 Mg of dolomite were applied in Korea (GIR, 2015). When limestone contacts strong acid sources in the soil, a chemical reaction is triggered, and some of the limestone degrades and releases CO₂ gas.

The CO₂ emission coefficients were proposed by the Intergovernmental Panel on Climate Change (IPCC) as 0.13 and 0.12 Mg C per Mg for dolomite and limestone, respectively (IPCC,

2006). These IPCC emission coefficients are the mass fractions of carbon (C) in two main liming materials and in the annual Inventory of Greenhouse Gas emissions, and sinks are currently used in most countries. However, the estimation that the entire C is released to the atmosphere as CO₂ from liming materials appears improbable based on current understanding of the very low water solubility of limestone and dolomite and carbonate transport through soil. The dissolution rate of applied liming materials depends on the soil properties, climate condition, and the type of mineral applied (EPA, 2014).

Carbon dioxide emissions from dissolved liming materials could be estimated with more detailed measurements that integrate the potential of bicarbonate leaching loss or precipitation with counter cations,

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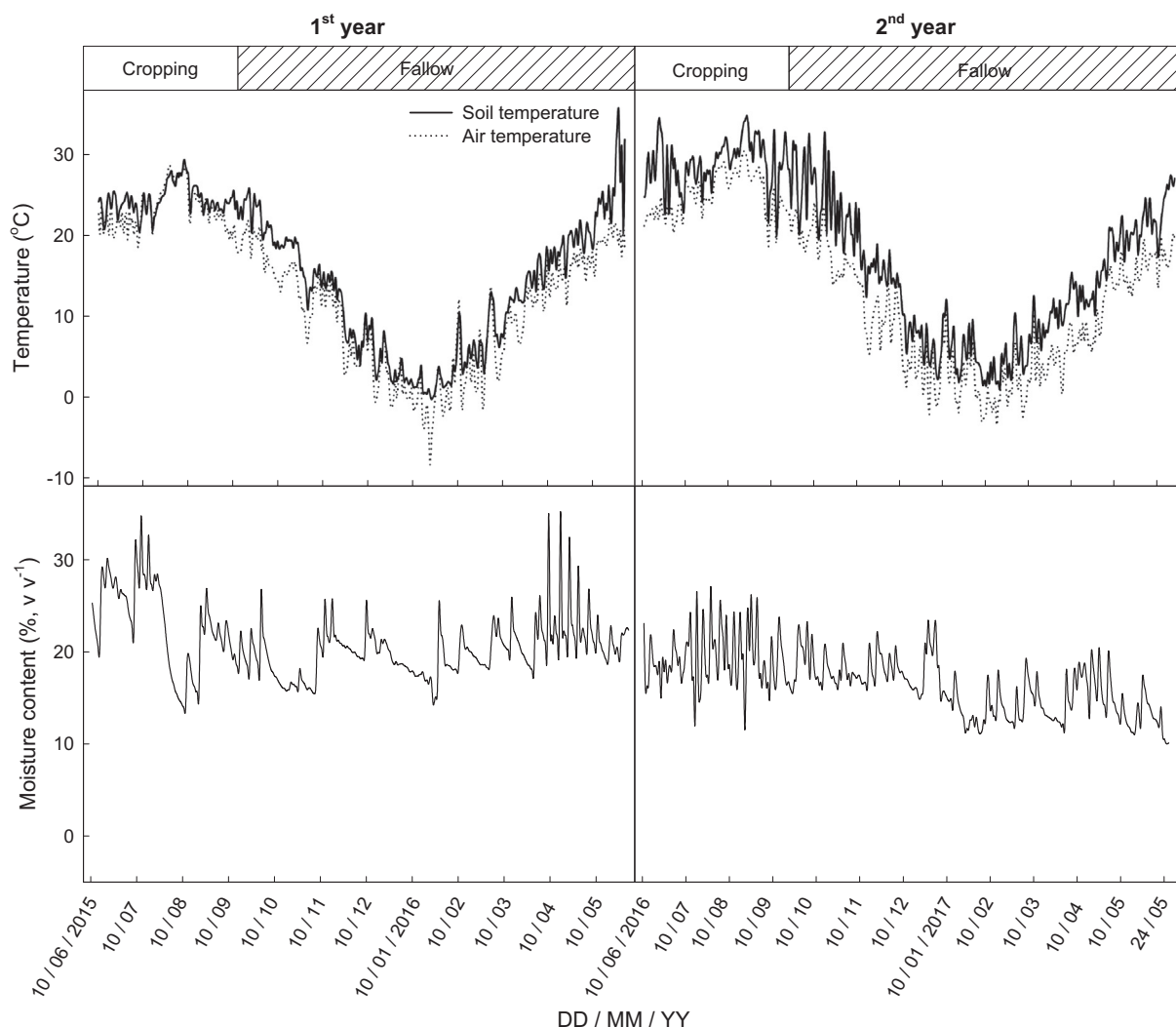


Fig. 1. Changes in soil, air temperature and soil moisture content.

thereby not immediately contributing to CO₂ emissions. However, the specific CO₂ emission factor from the applied liming materials was, until now, not developed for arable soils.

Therefore, this two-year field study was conducted to provide the direct CO₂ emission factor from C-containing liming materials applied in moderately acidic soil. Limestone (CaCO₃) was applied in a typical temperate upland soil at different levels (0–2 Mg ha⁻¹ a year), and CO₂ emission rates were annually monitored using the closed-chamber method. Finally, the direct CO₂-C emission factor from CaCO₃ was estimated using ¹³C-CO₂-C fluxes released from ¹³C-CaCO₃.

2. Materials and methods

2.1. Investigation plot installation

Investigation plots were installed in a typical temperate upland soil at the agronomy field of Gyeongsang National University (35°08'59 N, 128°05'47 E), Jinju, Kyeongnam, South Korea for two years (2015 and 2016). The soil was classified as the Daegu series (loamy skeletal, mixed, mesic family of lithic dystrudepts). The initial soil properties were pH 5.6 (1:2.5 with H₂O), total carbon content 23.7 g C kg⁻¹, total nitrogen content 2.4 g N kg⁻¹, electrical conductivity (EC) 0.63 dS m⁻¹, and exchangeable calcium (Ca²⁺) 5.2 cmol⁺ kg⁻¹. Lime application for maize is suggested when soil pH is < 6.5 (RDA, 2017). Four different levels of CaCO₃ (0, 0.5, 1.0, and 2.0 Mg ha⁻¹) were installed as

treatments based on the recommended level of liming (2.0 Mg ha⁻¹) in Korean upland soil. Each experimental plot (100 m²) was established with a randomized block design with 3 replications. A soil bank was placed between each treatment to create buffer zones (0.4 m) and to minimize nutrient mixing effects.

2.2. Maize cultivation

Maize (*Zea mays* L.) seedlings (two weeks old) were transplanted on early June 2015 and 2016 with a spacing of 30 × 30 cm. All the experiment treatments were applied in chemical fertilizers with the same levels of N-P₂O₅-K₂O (186–35–74 kg ha⁻¹), following the recommended dose for Korean maize cultivation (RDA, 2017). Urea for N, superphosphate for P₂O₅ and K₂O were used throughout the experiments. 50% of the total N, 100% of the total P₂O₅ and K₂O were applied to a basal fertilizer one day before transplantation, and another 50% of N fertilizer was applied on the 40th day after maize transplanting.

Maize was cultivated with natural precipitation. Grasses in the plot were properly hand-removed during cropping seasons. Maize plants were harvested in early September and the ears, stover and grain were manually separated and were oven-dried at 60 °C for 72 h, and their yield was measured. The soils were maintained without any cropping during the fallow seasons.

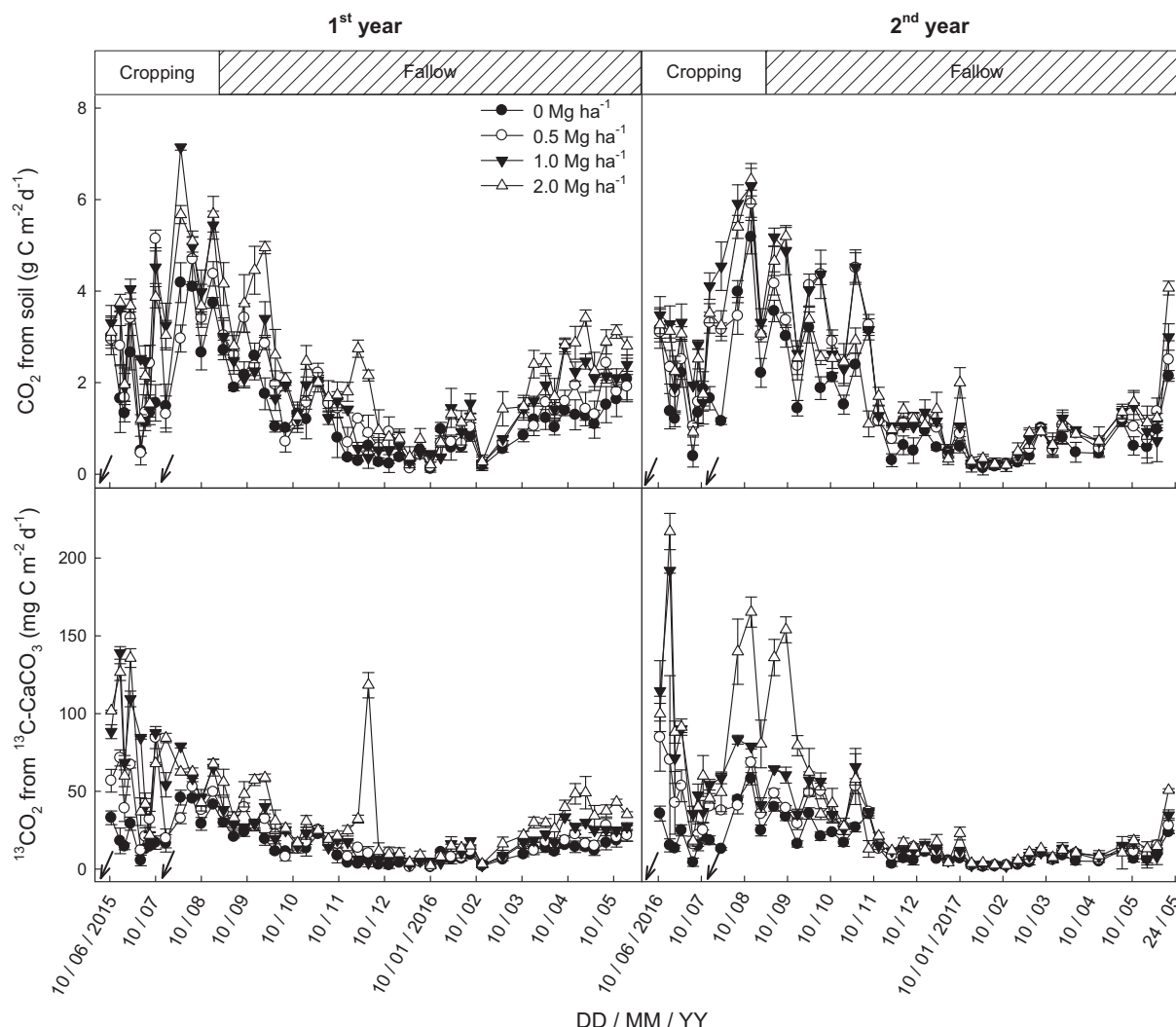


Fig. 2. Changes in CO_2 emission rates from soils and $^{13}\text{C}\text{-CaCO}_3$ under different levels of $^{13}\text{C}\text{-CaCO}_3$ application.

2.3. Investigation of CO_2 emission characteristics

Carbon dioxide flux in CaCO_3 applied soil was measured with the closed-chamber method during the entire experimental periods. An abiding cylindrical acrylic base chamber was buried in the ground (extending 7 cm above from the soil surface) with a hollow ring attached to its brim and installed in three replicates for each plot between maize plants, excluding maize roots (Jennifer et al., 2015). Gas samples were collected in a base chamber by covering it with opaque cylindrical acrylic chambers (25×24 cm). To prevent gas leaks during gas sampling, water was filled in the hollow ring on the brim. All of the chambers remained open during the whole experimental period, except during gas sampling in the field. The maize root intrusion was prohibited, and grasses inside the chamber were properly removed.

Inside the base chamber in each treatment, the same level of $^{13}\text{C}\text{-CaCO}_3$ (Sigma Aldrich, 99.99%) was applied to determine the emission rates of CO_2 induced from CaCO_3 . Gas samples were collected once a week in triplicates after closing the chamber manually using 30-mL volume airtight plastic syringes at 0 and 30 min intervals (Haque et al., 2015). However, gas sampling was conducted at two-day intervals for 2 weeks after $^{13}\text{C}\text{-CaCO}_3$ application to increase the accuracy. The collected gases were transferred into 20-mL volume air-evacuated glass vials.

To separate the gaseous components in samples, a Shimadzu QP 2010 plus GC/MS system equipped with a capillary silica column (SH-

Rt-Q-BOND, $30 \text{ m} \times 0.32 \text{ mm} \times 10 \mu\text{m}$ film thickness) in the splitless mode was used. The GC/MS detector operated at 1.10^{-5} Torr and 70 eV. Using the single ion monitoring mode, the detector could simultaneously quantify $^{12}\text{CO}_2$ ($m/z = 44$) from $^{13}\text{CO}_2$ ($m/z = 45$). The GC oven program was isothermal at 120°C . Carbon dioxide quantification was based on standards (Supelco Inc., Bellefonte, Pa.) and the serial dilutions prepared therefrom.

The CO_2 emission rate was estimated from the increase in CO_2 concentration per unit surface area of the chamber for a specific time interval by the following equation (Rolston, 1986; Lou et al., 2004).

$$R = (\Delta c / \Delta t) \times (V / A) \times \rho \times (273 / T)$$

where R: CO_2 emission rate ($\text{g m}^{-2} \text{d}^{-1}$); $\Delta c / \Delta t$: rate of CO_2 increase in the chamber ($\text{g m}^{-3} \text{d}^{-1}$); V: volume of the chamber (m^3); A: surface area of chamber (m^2); ρ : gas density of CO_2 under a standardized state (1.977 mg cm^{-3}); and T (absolute temperature): $273 + \text{mean temperature } (^\circ\text{C})$ of the chamber.

The total CO_2 flux was computed by the following equation (Singh et al., 1999).

$$\text{Total } \text{CO}_2 \text{ flux} = \sum^n (R_i \times D_i)$$

where R_i : CO_2 emission rate ($\text{g m}^{-2} \text{d}^{-1}$) in the i^{th} sampling interval; D_i : number of days in the i^{th} sampling interval; and n: number of sampling.

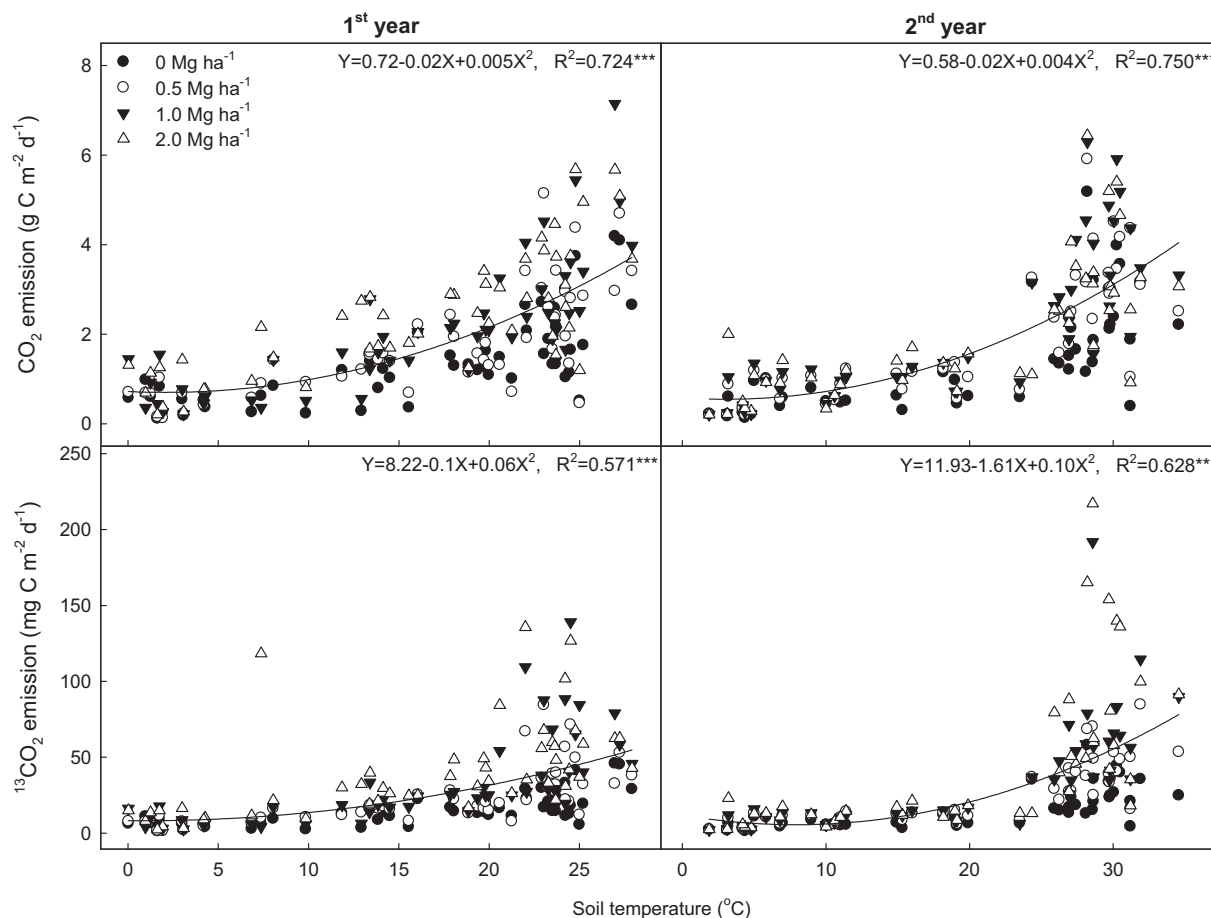


Fig. 3. Relationship between CO₂ emission rates and soil temperature under different levels of the ¹³C-CaCO₃ application.

2.4. Estimation of CO₂ emission from applied CaCO₃

Interpolation of ¹³CO₂ production based on the fixed ratio of ¹³C to C in naturally occurring C pools was selected to estimate the background ¹³CO₂ production (Padmanabhan et al., 2003). It is known that the natural abundance of ¹³C in nature is approximately 1.11% (Hoefs, 1987). In the 1st study year, this natural abundance rate was verified by correlating the CO₂ and ¹³CO₂ emission rates in the control treatment (Supplementary material-1 & 2). A very high linear correlation ($r = 0.999^{***}$, slope = 0.0116) was observed between CO₂ ($m/z = 44 + 45$) and ¹³CO₂ ($m/z = 45$), which means that the CO₂ emission derived from soil organic matter decomposition includes 1.16% of ¹³CO₂ emission. This ratio was used to estimate the background ¹³CO₂ production from ¹³C-CaCO₃ amended soils.

The naturally produced ¹³CO₂ flux was estimated by multiplying the natural abundant ¹³CO₂/CO₂ ratio (1.16%) and the CO₂ flux from ¹³C-CaCO₃ applied soils. Differences between the natural abundant ¹³CO₂ flux and the total ¹³CO₂ flux was used to assume the CO₂ flux from the ¹³C-CaCO₃ (Kim et al., 2016).

2.5. Analysis of soil and statistical property

The temperature and moisture content of soils were automatically recorded using sensors (5 TM Soil Moisture and Temperature sensor, Decagon Devices, WA, USA) at the surface soil (3–5 cm depth) throughout the whole investigation period. During the harvest stage, soils were collected with augur for chemical analysis. The collected soil was air-dried for 7 days and sieved with a 2 mm sieve. Chemical properties of soils were determined as follows: pH and electrical conductivity (1:2.5 with H₂O) and exchangeable cation (1 M ammonium

acetate, pH 7.0).

Statistical analyses were carried out in the SAS software package 9.1 (SAS Institute, 2003). Differences between years and lime application level parameters were determined using a two-way analysis of variance (ANOVA).

3. Results

3.1. Changes in the soil temperature and moisture content

Typical climatic conditions of the selected region were observed without a big variation in the two-year field studies. The overall changes in the soil moisture contents and temperatures did not significantly differ throughout the two-year investigation period (Fig. 1). The investigated two soil properties were not significantly different among treatments. However, very large differences were observed between seasons. For example, the mean soil temperatures during the cropping season were 23.5–25.1 °C, which were much higher than 9.6–10.2 °C during the fallow season. This tendency was similar to the soil moisture content changes. For example, the mean moisture content values during the cropping season were 23 and 20% ($v v^{-1}$) in the 1st and 2nd year, respectively, which were higher than 20 and 16% during each fallow season.

3.2. Changes in the soil respiration (CO₂ emission) rates

Irrespective of liming rates, typical CO₂ emission patterns were observed for two-year investigations (Fig. 2). Carbon dioxide emission rates were similarly changed with the change in soil temperature (Fig. 3). The emission rates increased with the rise in soil temperature

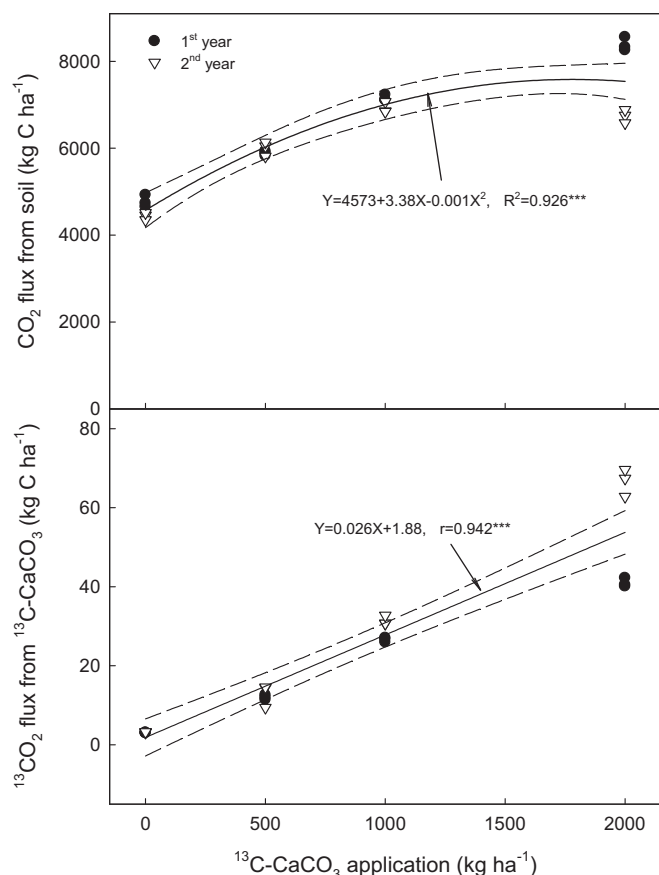


Fig. 4. Changes in the annual CO₂ fluxes from soil and ¹³C-CaCO₃ under different levels of the ¹³C-CaCO₃ application. (Note: dash lines are 95% of confidence).

and decreased with the decrease in soil temperature. The mean CO₂ emission rates were higher at 2.26–3.70 g CO₂-C m⁻² d⁻¹ during 90 days of the warm maize cropping season than at 0.92–1.94 g CO₂-C m⁻² d⁻¹ during the fallow season.

Annual CO₂ fluxes on the control (no liming) ranged within 4.4–4.7 Mg CO₂-C ha⁻¹, but lime addition significantly increased annual CO₂ fluxes with a quadratic response (Fig. 4). These variations were not significantly different between years. For example, annual average CO₂ fluxes for two years were altered with the relationship of $Y = 4573 + 3.38X - 0.001X^2$ ($R^2 = 0.926^{***}$, where X and Y indicate the lime application level in kg ha⁻¹ and the annual CO₂ flux in kg CO₂-

Cha⁻¹, respectively) (Fig. 4). The utmost annual CO₂ flux could be acquired at approximately 1900 kg ha⁻¹ of lime application. Thereafter, the CO₂ fluxes decreased with the increase in liming.

3.3. Emission of CO₂ from applied CaCO₃

The emission rates of CO₂ gas from ¹³C-CaCO₃ applied soils were similarly changed with CO₂ emission patterns (Fig. 2). The CO₂ emission rate changes were also closely associated with soil temperature changes (Fig. 3). Approximately 70–80% of annual CO₂ flux from ¹³C-CaCO₃ was emitted during the warm maize cropping season. Thereafter, CO₂ emission from ¹³C-CaCO₃ fluctuated with low rates (< 0.16 g CO₂-C m⁻² d⁻¹) (Fig. 2).

Annual ¹³CO₂ fluxes coming from ¹³C-CaCO₃ significantly increased with increasing ¹³C-CaCO₃ application levels at the rate of 0.026 Mg CO₂-C Mg⁻¹ CaCO₃ (Fig. 4). This result indicates that nearly 0.026 could be the direct CO₂ emission factor for lime amended in cropping upland.

3.4. Maize yield and soil properties

Lime application significantly increased the pH, electrical conductivity, and exchangeable Ca concentration in soil. These improved soil properties significantly stimulated maize growth and yield properties (Table 1). However, grain and stover yield properties were not significantly different between years or the year × CaCO₃ application level. The grain yield at the control treatment was approximately 4.2 Mg ha⁻¹, and 2 Mg ha⁻¹ of lime application increased its productivity by 16–18% over the control. The stover yields were similarly changed with grain yields to lime application.

4. Discussion

The IPCC has suggested that 100% of C in agricultural limes is released as CO₂ into the atmosphere (EPA, 2016; Houghton et al., 1997; Bernoux et al., 2003). For example, 0.12 Mg C per Mg of limestone (CaCO₃) is proposed as a CO₂ emission coefficient, which is the mass fraction of C in CaCO₃. However, the hypothesis that all C in CaCO₃ is expelled to the atmosphere as CO₂ gas is unlikely based on our synthesis of global carbonate (CO₃²⁻) dynamics in territories and in the ocean. In contrast, if the CaCO₃ was simply dissolved at one point and re-precipitated elsewhere, there should be no net emission of CO₂ (West and McBride, 2005).

Addressing the fate of C in liming materials during dissolution requires understanding the fractions of dissolution caused by carbonic acid (H₂CO₃) and strong acids (i.e., HNO₃). First, the CaCO₃ applied to soils can be dissolved into calcium (Ca²⁺) and bicarbonate (2HCO₃⁻)

Table 1

Soil chemical properties & maize yields under different levels of ¹³C-CaCO₃ application at harvesting stage.

Year (A)	CaCO ₃ application (kg ha ⁻¹) (B)	pH (1:5, H ₂ O)	Electrical conductivity (dS m ⁻¹)	Exchangeable cations (cmol ⁺ kg ⁻¹)				Grain yield (Mg ha ⁻¹)	Stover yield (Mg ha ⁻¹)
				Ca	Mg	K	Na		
1st	0	6.8	0.6	6.3	1.8	0.7	0.03	4.23	4.77
	500	7.1	0.7	6.7	1.8	0.9	0.01	4.59	4.89
	1000	7.0	0.9	6.8	1.6	1.0	0.01	4.71	4.98
	2000	7.3	0.9	7.2	1.8	0.8	0.10	4.90	5.46
2nd	0	6.7	0.7	6.4	1.9	0.8	0.05	4.26	4.64
	500	6.9	0.8	7.0	2.0	0.6	0.01	4.66	4.90
	1000	7.5	1.2	7.7	1.8	0.7	0.06	4.97	4.99
	2000	7.7	1.3	8.2	1.7	0.8	0.09	5.02	5.83
Statistical analysis									
A		***	NS	***	NS	NS	NS	NS	NS
B		***	NS	***	NS	NS	NS	*	*
A × B		***	NS	***	NS	NS	NS	NS	NS

(Note) ns, and *, ** and *** denote not significant, and significance at the 5, 1 and 0.1% levels, respectively.

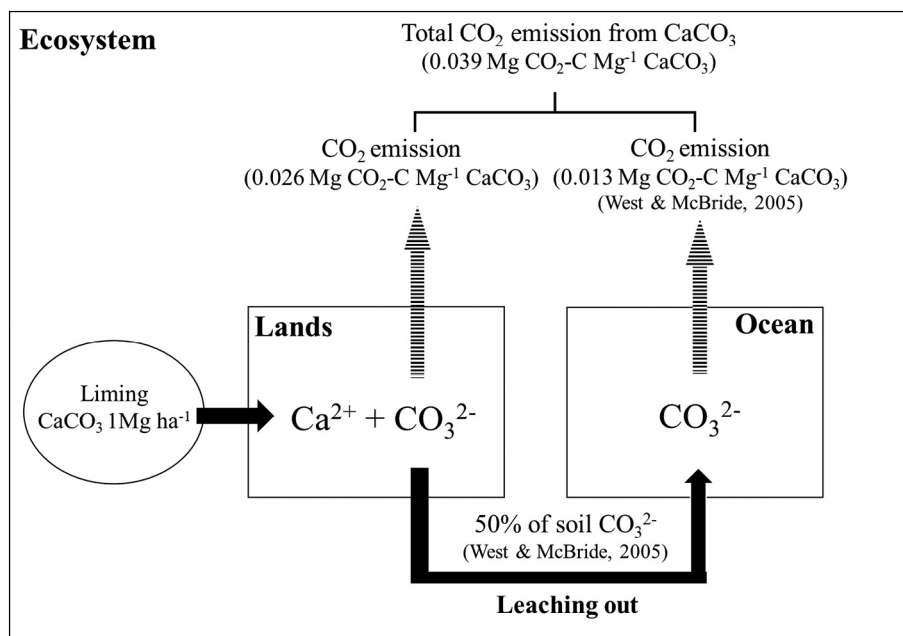
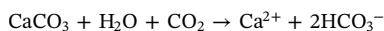
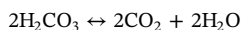


Fig. 5. The diagram of CO₂ flux from applied CaCO₃ in ecosystem.

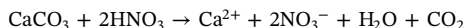
ions.



This reaction in soil solution produces 2 mol of HCO₃⁻ for every mole of gaseous CO₂ taken up. The HCO₃⁻ may either be flushed out or remain in the soil profile. It is expected that bicarbonate that remains in the soil reacts with available hydrogen (H⁺) and forms carbonic acid (H₂CO₃). This H₂CO₃ can convert into CO₂ and be released into the atmosphere. This reaction can release 1 mol of CO₂ for every mole of CaCO₃ dissolved.



On the other hand, the ammonium (NH₄⁺)-based nitrogen (N) fertilizer (i.e., urea and ammonium sulfate) and organic Ns can produce nitrate (NO₃⁻) and H⁺ via a nitrification process and form nitric acid (HNO₃). If CaCO₃ reacts with HNO₃, CO₂ can be formed. One mole of CO₂ can be released for every mole of CaCO₃ dissolved.



In determining the fraction of CO₂ emission from CaCO₃ applied, the USDA and the EPA have adopted different approaches. The USDA has considered agricultural CaCO₃ as a net C sink (−0.147 g CO₂ per g of CaCO₃), but the EPA has regarded it as a net CO₂ emission source (0.059 g C per g of CaCO₃). The USDA assumed that only one-third of CaCO₃ is acidified directly to CO₂ and that two-thirds of CaCO₃ is dissolved in the presence of H₂CO₃. The resultant H₂CO₃ will transport to the ocean and precipitate as CaCO₃ on a long-term basis. The USDA approach leads to a net C sink from the addition of CaCO₃ because half of the C in the H₂CO₃ comes from the uptake of atmospheric CO₂ (USDA, 2014). However, the USDA approach lacks resolution at the farm scale because the estimation was based on stream-monitoring data that were collected at the watershed scale. The USDA does not clarify the scientific basis that supports their assumption on the relative magnitudes of CaCO₃ that are acidified into CO₂ emissions and then into H₂CO₃, which has a direct impact on the estimated overall CO₂ emissions from agricultural liming.

In contrast, the EPA has used the findings that the net CO₂ emission from agricultural lime is 0.059 g C per g of CaCO₃ (EPA, 2016). This value was developed by compiling data on the use, dissolution, and transport of agricultural lime (West and McBride, 2005). The EPA

estimates that approximately 38% of CaCO₃ applied as a liming material is dissolved by HNO₃ and that this portion of input C is released as CO₂. The other 62% of CaCO₃ is dissolved by H₂CO₃ and results in the uptake of 1 mol CO₂ for every mole of CaCO₃ (third equation). Therefore, the C input and output is balanced in this pathway. Leaching and the transport of HCO₃⁻ and associated cations are the most uncertain factors in the EPA analysis. Using a 50% transport factor, 0.6 mol of CO₂ emission was estimated for every mole of CaCO₃ precipitated into the ocean (Ware et al., 1991; Sundquist, 1993). From this scenario, the EPA proposed 0.059 Mg C per Mg CaCO₃ as the net loss of CO₂ (West and McBride, 2005).

However, in the current field study in which ¹³C-CaCO₃ was applied in moderately acidic soil at different levels (0–2 Mg ha⁻¹) and the ¹³CO₂ emission factor from ¹³C-CaCO₃ was developed using annual ¹³CO₂ fluxes for two years, the direct CO₂ emission factor from CaCO₃ was 0.026 Mg C per Mg of CaCO₃ in a typical upland soil (Fig. 4). This value is approximately half of the CO₂ emissions from CaCO₃ that were estimated by EPA (2014). This difference might come from a different analysis boundary. In our field study, the CO₂ emission factor from CaCO₃ applied as a liming material was investigated only within the agricultural field boundary and was not included in the CO₂ emission from the CaCO₃ transported and precipitated into the ocean. In the EPA scenario, the direct CO₂ emission factor from CaCO₃ within the agricultural field was estimated with approximately 0.046 Mg C per Mg of CaCO₃ (West and McBride, 2005), which is approximately 2 times higher than our current CO₂ emission factor (0.026 Mg C per Mg of CaCO₃). In addition, the EPA proposed approximately 0.013 Mg C per Mg of CaCO₃ applied as the CO₂ emission factor in the ocean. If this EPA emission factor in the ocean could be added to the current CO₂ emission factor of the inner agricultural field, the whole CO₂ emission factor from lime could be close to 0.039 Mg C per Mg of CaCO₃ in temperate upland soil (Fig. 5).

In Korea, 46 and 193 thousand Mg of limestone and dolomite were applied as a soil amendment in 2013, respectively (GIR, 2015). This liming can emit a total 30.61 thousand Mg CO₂-C (5.52 and 25.09 thousand Mg CO₂-C from limestone and dolomite, respectively) using the IPCC default value (0.12 and 0.13 Mg C per Mg of limestone and dolomite, respectively). However, this estimation might be big different with the real CO₂ emission from liming materials applied in cropping lands. In this study, the CO₂ emission factor from dolomite was not

evaluated, but it might not be different with limestone, due to similar chemical properties. Total CO₂ emission from applied liming materials might be near 1.196 and 1.794 thousand Mg CO₂-C using the currently developed CO₂ emission factor from the inner cropping field (the direct CO₂ emission factor: 0.026 Mg C per Mg of limestone) and the whole ecosystem (0.039 Mg C per Mg), respectively.

5. Conclusion

The IPCC proposed 0.12 Mg C per Mg for limestone (CaCO₃) as the CO₂ emission coefficient, which indicates that 100% of C in agricultural liming materials is released as CO₂ into the atmosphere. However, we confirmed that 0.026 Mg C per Mg of CaCO₃ was annually emitted as CO₂ from agricultural liming material CaCO₃ in a moderately acidic soil. This value was slightly lower than the CO₂ emission factor of the EPA (0.059 Mg C per Mg of CaCO₃), which was combined with 0.046 Mg C per Mg of CaCO₃ and 0.013 Mg C per Mg of CaCO₃ of the CO₂ emission factor from the agricultural field and in the ocean, respectively. If the EPA emission factor from the ocean is added to the currently developed CO₂ emission factor inner the agricultural field, the total CO₂ emission factor may be close to 0.039 Mg C per Mg of CaCO₃ in the temperate upland soil. While more research is required to assay the loss of CO₂ coming from agricultural liming materials, our field investigation strongly suggests that not all C added to soil as liming materials is emitted into the atmosphere as CO₂.

Acknowledgements

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.geoderma.2018.10.007>.

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